Nature Physical Science

December 4, 1972, Vol. 240, No. 101, pp. 99-101 There is not a 1:1 correspondence of page numbers here with those in the published manuscript.

Xenon in Carbonaceous Chondrites

O. K. MANUEL & E. W. HENNECKE Nuclear Division, Department of Chemistry, University of Missouri, Rolla, Missouri 65401

D. D. SABU Department of Chemistry, Grambling College, Grambling, Louisiana 71254

Xenon in Carbonaceous Chondrites

Carbonaceous chondrites contain two isotopically distinct components of trapped xenon which cannot be explained by the occurrence of nuclear or fractionation processes within these meteorites.

Reynolds and Turner¹ reported enrichment of the heavy xenon isotopes in the gases released from the carbonaceous chondrite, Renazzo, at 700°-1,000° C. Subsequent studies of xenon in other carbonaceous chondrites have confirmed the existence of a "Renazzo-type" anomaly²⁻⁷, and it has been suggested that the excess heavy xenon isotopes may result from the spontaneous fission of ²⁴⁴Pu (ref. 1), from neutron-induced fission², from a "carrier" of heavy xenon adsorbed on carbonaceous material⁵, from fission of superheavy elements⁸⁻¹¹, or from mass fractionation¹².

In reviewing the available data on xenon released from carbonaceous chondrites by stepwise heating, we recently noticed an enrichment of the light xenon isotopes in the temperature fractions ($\approx 600^{\circ}$ -1,000° C) which also show an enrichment of the heavy xenon isotopes. Table 1 shows isotopic data for carbonaceous chondrites at those extraction temperatures showing the most spectacular enrichments of the heavy xenon isotopes. The isotopic composition of xenon in air¹³ and that of the total trapped xenon in average carbonaceous chondrites^{14, 15} (AVCC) are shown for comparison.

Table 1 Xenon Isotopes in Carbonaceous Chondrites									
Sample	124	126	128	130	131	132	134	136	References
Air	2.353	2.206	47.03	100	519.1	659.1	255.9	217.4	Nier ¹³
AVCC	2.828	2.533	50.38	100	506.7	620.9	236.9	199.0	Marti ¹⁴ and Eugster et al. ¹⁵
Renazzo 800°	2.93 ± 0.05	2.57 ± 0.03	50.6±0.4	100	505.8± 2.4	617.2± 1.9	243.3 ± 2.2	210.4 ± 1.5	Reynolds and Turner ¹
Leoville 1,010°	3.07 ± 0.09	2.66 ± 0.12	51.4±0.5	100	511.3± 4.9	625.0± 3.9	250.0 ± 2.4	220.6±2.3	Manuel et al.6
Makoia (matrix) 600°	3.15 ± 0.09	2.72 ± 0.09	52.7±0.7	100	519.9± 6.8	633.3± 7.6	5 276.1 ± 3.6	248.9±3.6	Rowe ⁵
Makoia 750°	3.39 ± 0.06	2.90±0.05	52.0 ± 0.8	100	514.1± 5.6	624.5± 5.6	278.6±3.2	261.1 ± 2.0	Manuel et al.7
Allende 1,000°	3.39 ± 0.14	2.77 ± 0.09	54.5 ± 0.9	100	513.0± 6.1	624.9± 8.6	291.2±3.9	282.5 ± 2.4	Manuel et al.7
Allende 800°	3.62±0.13	2.84±0.09	52.1±1.5	100	518.6±11.9	641.0±14.3	305.1±7.0	299.4±5.4	Manuel et al. ⁷

In Table 1 the xenon from carbonaceous chondrites is shown in the order of increasing enrichments of the heavy xenon isotopes. This arrangement of the xenon data also corresponds to an increasing enrichment of the lighter xenon isotopes. The absence of any significant spallation contribution to the light xenon isotopes in the "Renazzo-type" anomaly is indicated by the fact that all of the observed 124 Xe/ 126 Xe ratios are higher than that in AVCC xenon, whereas all known spallation reactions yield 124 Xe/ 126 Xe ratios ${}^{16, 17}$ that are lower than the 124 Xe/ 126 Xe ratio in AVCC xenon.



Fig. 1 Correlation observed between enrichments of 124 Xe/ 130 Xe ratios and 136 Xe/ 130 Xe ratios in carbonaceous chondrites which exhibit the "Renazzo-type" anomaly. The isotope data for air (*A*); AVCC (*V*) and solar (*S*) xenon are from Nier¹³, Marti¹⁴, Eugster *et al.*¹⁵, and Kaiser²⁰, respectively, and the isotope data from stepwise heating of carbonaceous chondrites are from Reynolds and Turner¹, Manuel *et al.*⁶, Rowe⁵ and Manuel *et al.*⁷.

Fig. 1 shows the observed correlation of the 124 Xe/ 130 Xe ratios with the 136 Xe/ 130 Xe ratios in these carbonaceous chondrites and the correlations expected from mixtures of AVCC xenon with products from fission or from spallation reactions. The pattern expected from mass dependent fractionation is also shown. It appears from the data in Table 1 and Fig. 1 that the observed correlation cannot be accounted for by any of the previously proposed mechanisms. Fission induced by bombardment at high energies yields both neutron- rich and neutron-poor isotopes of xenon, but it has been shown¹⁸ that the yields of the neutron-poor isotopes from this process are like those from spallation reactions – high energy induced fission produces a 124 Xe/ 126 Xe ratio smaller than that in AVCC xenon. We know of no plausible in situ nuclear process which could produce the observed enrichment of the lightest and heaviest isotopes of xenon. Further, the high degree of correlation of ¹²⁴Xe/¹³⁰Xe with ¹³⁶Xe/¹³⁰Xe (98%) seems to rule out any accidental mixture of in situ produced enrichments of the heaviest and the lightest isotopes. We therefore conclude that the "Renazzo-type" anomaly is due to an isotopically distinct component of xenon (hereafter referred to as X) which has been incorporated into the carbonaceous chondrites. The correlations for the xenon isotope ratios seen in Table 1 and Fig. 1 are then the result of mixtures of AVCC xenon with this component **X**.

It has been suggested that atmospheric and solar-wind implanted xenon are related by isotopic mass fractionation^{19, 20}. The dashed line through atmospheric xenon in Fig. 1 shows the effect expected from Aston's equation²¹ for diffusive fractionation. Also shown is a recent estimate of the isotopic composition of solar-wind implanted xenon obtained from analysis of lunar soils²⁰. The shift of AVCC xenon from the fractionation line relating atmospheric and solar xenon suggests that AVCC xenon may contain a small component of X.

Properties of Anomalous Xenon

The xenon released from Allende at 800° C contains a higher percentage of **X** than does any other xenon reported to date. We therefore use this to seek information on the relative abundances of the isotopes and possible origin of component **X**. From the data shown in Table 1 it appears that **X** shows the greatest enrichment in the lighter isotopes, ^{124, 126}Xe, and the heavier isotopes, ^{134, 136}Xe, relative to ¹³⁰Xe. These are the isotopes expected to be produced in supernova explosions²², the ^{124, 126}Xe isotopes by the p-process and the ^{134, 136}Xe by the r-process. Thus, it is possible that **X** in carbonaceous chondrites represents material that has been added to our solar system from a nearby supernova, although no evidence for the addition of products from a separate nucleosynthesis event has been found in the other elements.

To look for a more quantitative relationship between **X** and xenon in the Earth's atmosphere, we employ the method of Canalas *et al.*²³ to compute a fractionation factor, F^{m} , for each xenon isotope.

$$F^{m} = ({}^{i}Xe/{}^{130}Xe)_{Met}/({}^{i}Xe/{}^{130}Xe)_{Air}$$
(1)



Fig. 2 A comparison of the abundance pattern of xenon isotopes from the 800° fraction of Allende⁷ relative to the abundance of xenon isotopes in the atmosphere¹³. The dashed line shows the mass fractionation effects calculated from Aston's equation²¹ for diffusive gas loss.

Fig. 2 shows the values of F^m for the xenon isotopes released from Allende at 800° C. The values of F^m for the light isotopes, ¹²⁴⁻¹³⁰Xe, decrease in a regular manner with increasing mass number, but the F^m values for the heavier isotopes, ¹³¹⁻¹³⁶Xe, show an irregular pattern with increasing mass number. The pattern seen across the lighter xenon isotopes is suggestive of mass dependent fractionation, a process which has recently been recognized as the source for many isotopic anomalies of noble gases^{6, 7, 12, 24-26}. The dashed line in Fig. 2 shows the pattern expected across the fractionation factors, F^m , if

atmospheric xenon and **X** are related by the diffusive gas loss equation of Aston²¹. The dashed line corresponds to a volume change in xenon of $\approx 3 \times 10^7$. The abundance pattern of the lighter isotopes, ¹²⁴⁻¹³⁰Xe, fits Aston's equation and there remains an excess of the heavier xenon isotopes in the ratio ¹³¹Xe: ¹³²Xe: ¹³⁴Xe: ¹³⁶Xe = 0.30 ± 0.11 : 0.58 ± 0.14 : $\equiv 1.00$: 1.41 ± 0.10. Thus, the correlation shown in Fig. 2 suggests that the **X** that has been incorporated into carbonaceous chondrites can be related to atmospheric xenon by a combination of isotopic mass fractionation plus some process which produced the heavy xenon isotopes in the above ratio.

Possible Sources of X

The suggestion that meteoritic and atmospheric xenon may be related by a mixture of fission and fractionation dates back to Reynolds's discovery²⁷ of anomalous meteoritic xenon in 1960. This view is widely accepted, and several investigators^{1-5, 14, 15, 28, 29} have noted that the xenon in carbonaceous chondrites may contain a fission component with yields similar to the excess heavy xenon isotopes calculated above for the 800° fraction of Allende. Earlier workers have not noticed the correlation of excess heavy and light xenon isotopes, which suggests that the anomalies of **X** were not produced *in situ* but were present when the xenon was incorporated into the meteorites.

Because **X** and solar-wind implanted xenon are released from meteorites at about the same extraction temperatures¹⁹, it appears that **X** may have been implanted from an early solar wind. The xenon implanted by the solar wind in lunar soils is not as enriched in the light isotopes as is X. Changes in the fractionation of noble gas isotopes in the solar wind may be expected, however, because there is evidence for time variation in the degree of fractionation of gases presently reaching the lunar surface³⁰.

The origin of excess ¹³⁰⁻¹³⁶Xe in **X** is more difficult to explain by current theories of the early solar system. Pepin² noted that the pattern of excess ¹³⁰⁻¹³⁶Xe in carbonaceous chondrites resembled that produced by a high flux of thermal neutrons on ²³⁵U, but he concluded that the absence of detectable neutron-capture anomalies in the rare earths of meteorites left no satisfactory explanation for the excess heavy xenon isotopes. A thermal neutron flux of $\approx 2.8 \times 10^{13}$ neutrons cm⁻² s⁻¹ on ²³⁵U would produce ¹³¹Xe : ¹³²Xe : ¹³⁴Xe : ¹³⁶Xe = 0.36 : 0.54 : 1.00 : 1.41, in agreement with the yields calculated above for **X**. As we have shown that this enrichment of heavy xenon isotopes is not produced by *in situ* nuclear processes, the absence of neutron-capture anomalies in the rare earths is no longer an obstacle to this model. Also because of the large thermal neutron capture crosssection of ¹³⁵Xe, a high thermal neutron flux would yield a large ¹³⁶Xe : ¹³⁴Xe ratio for the induced fission of any transbismuth nuclide. An early deuterium burning stage in the outer region of the Sun or the irradiation of planetary material prior to accretion into planetary bodies, as proposed by Fowler *et al.*³¹, are possible sources of the neutrons.

In summary, the enrichment of heavy xenon isotopes released from carbonaceous chondrites near 600° -1,000 C is accompanied by an enrichment of the light xenon isotopes. The high degree of correlation between these two isotopic anomalies suggests

that both result from a common source. Because no known nuclear or physical process is capable of producing both anomalies *in situ*, we suggest that they result from the release of isotopically anomalous xenon (component \mathbf{X}) that was trapped in the meteorites.

This investigation was supported by NASA.

Received August 14; revised October 27, 1972.

- 1. Reynolds, J. H., and Turner, G., J. Geophys. Res., 69, 3263 (1964).
- 2. Pepin, R. O., *Origin and Distribution of the Elements* (edit. by Ahrens, L. H.), 379 (Pergamon Press, Oxford and New York, 1968).
- 3. Turner, G., J. Geophys. Res., 70, 5433 (1965).
- 4. Funk, H., Podosek, F., and Rowe, M. W., *Geochim. Cosmochim. Acta*, **31**, 1721 (1967).
- 5. Rowe, M. W., Geochim. Cosmochim. Acta, 32, 1317 (1968).
- Manuel, O. K., Wright, R. J., Miller, D. K., and Kuroda, P. K., J. Geophys. Res., 75, 5693 (1970).
- 7. Manuel, O. K., Wright, R. J., Miller, D. K., and Kuroda, P. K., *Geochim. Cosmochim. Acta*, **36**, 961 (1972).
- Srinivasan, B., Alexander, E. C. jun., Manuel, O. K., and Troutner, D. E., *Phys. Rev.*, **179**,1166 (1969).
- 9. Dakowski, M., Earth Planet. Sci. Lett., 6, 152 (1969).
- 10. Anders, E., and Heymann, D., Science, 164, 821 (1969).
- 11. Rao, M. N., Nucl. Phys., A, 140, 69 (1970).
- 12. Kuroda, P. K., and Manuel, O. K., Nature, 227, 1113 (1970).
- 13. Nier, A. O., Phys. Rev., 79, 450 (1950).
- 14. Marti, K., Earth Planet. Sci. Lett., 3, 243 (1967).
- 15. Eugster, O., Eberhardt, P., and Geiss, J., Earth Planet. Sci. Lett., 3, 249 (1967).
- 16. Funk, H., and Rowe, M. W., Earth Planet. Sci. Lett., 2, 215 (1967).
- 17. Hohenberg, C. M., and Rowe, M. W., J. Geophys. Res., 75, 4205 (1970).
- Hudis, J., Kirsten, T., Stoenner, R. W., and Schaeffer, O. A., *Phys. Rev.*, **1C**, 2019 (1970).
- 19. Marti, K., Science, 166, 1263 (1969).
- 20. Kaiser, W. A., Earth Planet. Sci. Lett., 13, 387 (1972).
- 21. Aston, F. W., *Mass-Spectra and Isotopes*, 219 (Edward Arnold & Co., London, 1933).
- 22. Burbidge, E. M., Burbidge, G. R., Fowler, W. A., and Hoyle, F., *Rev. Mod. Phys.*, **29**, 547 (1957).
- 23. Canalas, R. A., Alexander, E. C. jun., and Manuel, O. K., J. Geophys. Res., 73, 3331 (1968).
- 24. Manuel, O. K., Meteoritics, 5, 207 (1970).
- 25. Hennecke, E. W., and Manuel, O. K., Z. Naturforsch., 26a, 1980 (1971).
- 26. Kuroda, P. K., Reynolds, M. A., Sakamoto, K., and Miller, D. K., *Nature*, **230**, 42 (1971).
- 27. Reynolds, J. H., Phys. Rev. Lett., 4, 8 (1960).
- 28. Reynolds, J. H., Phys. Rev. Lett., 4, 351 (1960).

- 29. Krummenacher, D., Merrihue, C. M., Pepin, R. O., and Reynolds, J. H., *Geochim. Cosmochim. Acta*, **26**, 231 (1962).
- 30. Geiss, J., Buehler, F., Cerutti, H., Eberhardt, P., and Filleux, Ch., *Solar-wind Composition Experiment*, Apollo 16 Preliminary Science Report (in the press).
- 31. Fowler, W. A., Greenstein, J. L., and Hoyle, F., Amer. J. Phys., 29, 393 (1961).

Printed in Great Britain by Flarepath Printers Ltd., St. Albans, Herts.

